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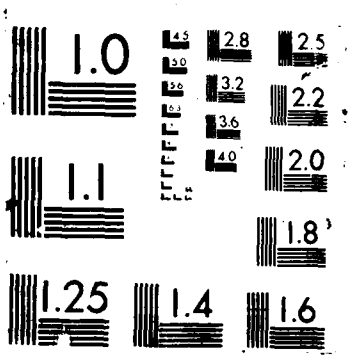
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Light Absorption by an Atom Moving Inside a Spherical Box

by

Isidore Last and Thomas F. George

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State University of New York at Buffalo
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19. ABSTRACT (Continue on reverse if necessary and identify by block number)

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Light Absorption by an Atom Moving Inside a Spherical Box

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Abstract

The radiative transition for an atom moving inside a spherical box is considered in the terms of electronic-field states. By using the hopping model, an analytical expression is obtained for the transition probability. This expression is compared with results obtained by numerical integration of the time-dependent Schrödinger equation.

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I. Introduction

When the energy of an electronic transition in an atom depends on its distance to another atom or molecule, then a collisional radiative transition is possible whose frequency is different from that of the free atom. Collisional radiative transitions have been considered theoretically for both atom-atom [1-5] and atom-molecule [6-11] systems. The absorption or emission of a photon in a colliding system is conveniently described as a dynamical transition between electronic-field states which are built from the field-free states by using the rotating-wave approximation [7,12-14]. The limitations of this approximation and consequently the limitations of the description of collisional radiative transitions in terms of electronic-field states have been analyzed.[15]

A situation similar to that of a collisional radiative transition exists in the case of guest atom inside a solid matrix, so far as the levels of the guest atom depend on its position $\vec{r}$ in the matrix. If the guest atom is moving, then a photon $\hbar\omega$ can be absorbed when the atom goes through a surface formed by points with transition energy $E_k - E_0$ equal to the photon energy,

$$E_k(\vec{r}_\omega) - E_0(\vec{r}_\omega) = \hbar\omega \quad . \tag{1}$$

The $\hbar\omega$ absorption can be considered as a transition between electronic-field states with the avoided crossing at the surface $\vec{r}_\omega$, in accordance with the theory of collisional radiative transitions. Assuming the host (matrix) atoms to be fixed, one describes the dynamics of the system as the semiclassical motion of the guest atom in a real three-dimensional space.

The thermal motion of an atom is of interest only for systems where the ground state energy $E_0(\vec{r})$ is not changing much inside some volume in the matrix. This is possible usually in a matrix containing relatively small guest atoms in

substitutional sites. An interesting example of such a system is represented by a xenon matrix containing Cl atoms which are obtained by photodissociation of Cl_2 or HCl molecules [16]. Since the Cl atom is smaller than a Xe atom, it has some freedom of motion inside a Xe cage [16-17]. This is supported by an experimental study of the radiative excitation of ionic states with electron transfer from the Xe matrix to the Cl atom [16]. According to semiempirical calculations performed by the diatomics-in-ionic states method [18], the ground state potential energy surface (PES) is almost flat inside a sphere with a radius of ~1 Å and increases sharply at distances larger than 1.1-1.2 Å from the cage center [19]. This PES can be approximated by the simple model of a spherical box with a rigid wall of infinite energy.

The study of the photoabsorption by an atom moving in such a spherical box is the subject of this paper. A model of a spherical box may be a satisfactory approximation to any system where an atom is located in a cage of high symmetry. The absorption of light by an atom in a spherical box is described here as a transition between two electronic-field states. This transition is considered both by using the Landau-Zener formula and by solving the time-dependent semiclassical Schrödinger equation.

II. Spherical Box Model

Let us consider an atom inside a spherical box with a radius R_0. The ground state potential energy V_1 is assumed to be zero inside the box and infinity outside it. Consequently, the atom is moving freely inside the box until it collides with the wall of the box. We shall consider only the thermal motion of the atom, where collisions with the box wall are suggested to be strong enough to change the direction and the velocity of the motion in a random way. The atom can be excited by a photon to an excited electronic state V_2 for

which the PES is spherically symmetric. The excitation may involve also the electrons of surrounding atoms, like in the case of a Cl atom in a Xe cage as described above. The excited state PES is suggested to have the simple parabolic form

$$V_2 = V_c - (V_c - V_0)R^2/R_0^2 \quad , \tag{2}$$

where V_c and V_0 are the energies in the box center ($R = 0$) and at the edge of the box ($R = R_0$), respectively. The excitation to the state V_2 is possible if the photon energy lies within the limits of the potentials V_0 and V_c,

$$V_0 \leq \hbar\omega \leq V_c \quad . \tag{3}$$

The radiative coupling u between the ground and excited states is assumed to be constant.

The radiative transition between two electronic states will be described here in terms of the electronic-field PES W_1 and W_2 [7]. Taking into account that the ground state field-free PES is zero ($V_1 = 0$), one obtains

$$W_{1,2} = \tfrac{1}{2}(V_2 + \hbar\omega) \pm \tfrac{1}{2}[(V_2 - \hbar\omega)^2 + 4u^2]^{\frac{1}{2}} \quad . \tag{4}$$

The electronic-field PES have an avoided crossing at the spherical seam whose radius, according to Eq. (2) is,

$$R_s = \left[\frac{V_c - \hbar\omega}{V_c - V_0}\right]^{\frac{1}{2}} R_0 \quad . \tag{5}$$

The radiative transition is described now as an adiabatic motion along the electronic-field PES W_1 or W_2, whereas the motion without radiative transition is described as diabatic motion with a nonadiabatic transition between the electronic-field PES in the region of the seam (5).

We shall assume the coupling terms to be small, so that the probability of the radiative transition between two successive collisions of the atom with the box wall is much smaller than unity. However, the coupling has to be strong enough to meet the criterion of the rotating-wave approximation [15]. In the present model this criterion is

$$\gamma = \frac{2\pi v|V_c - V_0|}{u\omega R_0} < 3 \quad . \tag{6}$$

In the case of a Cl atom in a Xe cage ($R_0 \approx 1$ Å, $V_c - V_0 \approx 0.4$eV, $\hbar\omega \approx 3$eV, $v \approx$ 100 m/s) the condition $\gamma = 3$ is fulfilled for a relatively small lower boundary of the coupling, $u_{min} \approx 2\times10^{-4}$ eV.

III. Transition Probability in the Landau-Zener Approximation

The probability of the radiative transition can be obtained by using the trajectory-hopping model [20-21] for the motion on electronic-field PES. In this model the transition is considered as a hopping between states which takes place when the trajectory crosses a seam, i.e., the crossing surface of the diabatic PES. The probability of hopping is calculated usually within the Landau-Zener approximation. In the present model the seam is a sphere with the radius given in (5). The trajectory, which has a form of a straight line, crosses the seam twice between two successive collisions with the box wall. The Landau-Zener transition probability for one crossing is

$$p = 1 - \exp[2\pi u^2/(\hbar|\frac{dV_2}{dn}|v_n)] \quad , \tag{7}$$

where $\frac{dV_2}{dn}$ is the component of the potential energy derivative normal to the seam and v_n is the normal component of the velocity. Taking into account that P is assumed to be much smaller than unity, as well as the geometry of the seam crossings (Fig. 1), one obtains the following expression for the transition probability per one run between the box walls:

$$p_\beta = \frac{4\pi u^2}{v\hbar|\frac{dV_2}{dR}|_s} \frac{\cos\beta_s}{[\cos^2\beta_s - \cos^2\beta]^{\frac{1}{2}}} \quad , \quad \beta < \beta_s \tag{8}$$

where $|\frac{dV_2}{dR}|_s$ is determined at the seam, β is the angle of motion, and β_s is the seam angle from the starting point (Fig. 1),

$$\beta_s = \sin^{-1}(R_s/R_0) \tag{9}$$

The transition probability per time unit is obtained as an average over the angle β,

$$P = \int_0^{\frac{\pi}{2}} d\beta\, p_\beta \sin\beta \,/ \int_0^{\frac{\pi}{2}} d\beta\, \tau_\beta \sin\beta \quad , \tag{10}$$

where τ_β is the time of one run for a fixed angle β,

$$\tau_\beta = 2R_0\cos\beta/v \quad . \tag{11}$$

Combining Eqs. (8), (10) and (11) and taking into account that $p_\beta = 0$ for $\beta > \beta_s$ and that u is constant, one obtains an analytical expression for the transition probability:

$$P = \frac{4\pi^2}{R_o \hbar \left|\frac{dV}{dR}\right|_s} \sin\beta_s \ln(\sec\beta_s + \tan\beta_s) \quad . \tag{12}$$

The transition probability (12) does not depend on the atom velocity v. In the case of the parabolic potential given by (2), the seam angle (9) is determined from the equation

$$\sin\beta_s = \frac{R_s}{R_0} = \left[\frac{V_c - \hbar\omega}{V_c - V_0}\right]^{\frac{1}{2}} \quad , \tag{13}$$

and the transition probability (12) becomes

$$P = \frac{u^2}{\hbar(V_c - V_0)} G(\beta_s) \quad , \tag{14}$$

$$G(\beta_s) = 2\pi \ln(\sec\beta_s + \tan\beta_s) \quad . \tag{15}$$

According to the last expression, the transition probability per time unit does not depend on the box radius R_0 and is inversely proportional to the potential difference $V_c - V_0$. Its dependence on the photon energy $\hbar\omega$ is expressed in a universal way as a function of the seam angle β_s [see Eq. (13)].

For small $\sin\beta_s$, when the photon energy is close to the transition energy in the box center V_c, the transition probability is proportional to $\sin\beta_s$ or to the square root of the difference $V_c - \hbar\omega$:

$$P \approx \frac{2\pi u^2}{\hbar(V_c - V_0)} \sin\beta_s = \frac{2\pi u^2 (V_c - \hbar\omega)^{\frac{1}{2}}}{\hbar(V_c - V_0)^{3/2}} \quad . \tag{16}$$

As the photon energy approaches the lower limit of the potential V_0, the transition probability increases to infinity as the logarithm of the difference $\hbar\omega - V_0$,

$$P \approx \frac{\pi u^2}{\hbar(V_c - V_0)} \left| \ln\left(\frac{\hbar\omega - V_0}{V_c - V_0}\right) \right| \tag{17}$$

As will be shown later, this increase of P to infinity for $\hbar\omega$ close to V_o is due to the Landau-Zener approximation.

IV. Transition Probability as the Solution of the Time-Dependent Schrödinger Equation

In the hopping model considered above, the transition takes place at the seam only with a probability determined by the Landau-Zener formula. These two assumptions are discarded within the framework of the "exact" semiclassical approach where the transition probability is determined by solving the time-dependent Schrödinger equation for the electronic states of an atom moving on classical trajectories [23]. The exact semiclassical transition probabilities may differ significantly from the Landau-Zener probabilities, as it was shown for the case of radiative transitions in atom-molecule collisions [24].

If the transition probability is much smaller than unity, as assumed above, then the amplitude of the final state, c_2, is determined by the integral [25]

$$c_2(\tau) = -\frac{i}{\hbar} \int_0^{\tau} dt\ u \exp\left[\frac{i}{\hbar} \int_0^{t} dt' \ (V_2 - \hbar\omega)\right] \quad . \tag{18}$$

For molecular collisions the transition probability is equal to the square of the amplitude c_2 at infinity. In the box model we shall assume that a collision with the box wall changes significantly the phase of the electronic states, and consequently the transition probability can be presented as a sum of the transition probabilites determined for separate free runs. This simplification allows us to express the transition probability per time unit in a similar way as in the Landau-Zener approximation [see Eq. (10)],

$$P = \int_0^{\frac{\pi}{2}} d\beta \, |c_2(\tau_\beta)|^2 \sin\beta \, / \int_0^{\frac{\pi}{2}} d\beta \, \tau_\beta \sin\beta \quad , \tag{19}$$

where τ_β is the free run time interval (11). Substituting the expression for the potential (2) into the integral (19), one obtains the coefficient c_2 as a function of the angle β,

$$c_2(\beta) = - \frac{iuR_0}{\hbar v} Q(\beta) \quad , \tag{20}$$

$$Q(\beta) = \int_0^{2\cos\beta} d\xi \, \exp(i\phi) \quad , \quad \xi = vt/R_0 \tag{21}$$

where ϕ is the phase,

$$\phi = f[\cos^2\beta_s + \cos\beta\xi - \tfrac{1}{3}\xi^2]\xi \quad , \tag{22}$$

$$f = \frac{R_0(V_c - V_0)}{v\hbar} \quad . \tag{23}$$

Substituting Eq. (20) into (19), one obtains the transition probability as a function of the seam angle β_s [see Eq. (13)],

$$P = \frac{u^2}{\hbar(V_c - V_0)} F_f(\beta_s) \quad , \tag{24}$$

$$F_f(\beta_s) = f \int_0^{\frac{\pi}{2}} d\beta \; |Q(\beta)|^2 \sin\beta \quad . \tag{25}$$

The exact semiclassical expression (24) is similar to the Landau-Zener expression (14) with the difference only in the dependence on the seam angle β_s. In the case of the exact semiclassical approach, the function F_f which describes the dependence on β_s includes one parameter, namely the unitless value (23), whereas in the Landau-Zener case the corresponding function G does not include any parameters at all.

V. Calculational Results

The results of the numerical calculation of the function F_f which determines the exact semiclassical transition probability (24) are presented in Fig. 2 for three values of the parameter f. According to these results, F_f does not depend on f within a wide range of seam angles β_s where it coincides with the function G of the Landau-Zener probability (14). When β_s is large, F_f first becomes a bit larger than the F function, but in the region close to 90° it begins to decrease whereas G increases to infinity. The smaller the parameter f is, the smaller is the angle β_s of the F_f maximum. The exact semiclassical transition probabilites per time unit for three atom velocities are presented in Fig. 3 together with the Landau-Zener probability.

The results of the calculation lead us to conclude that the transition probability for an atom in a spherical box with the potential (2) is determined by a simple analytical formula (14) within a wide interval of the seam angles β_s, for example $\beta_s \lesssim 70°$ for f = 75 and $\beta_s \lesssim 80°$ for f = 1200. These intervals of β_s cover 88% and 97% of the photon energy interval (3), respectively.

In order to describe analytically also the transition probabilities for $\hbar\omega$ close to the lower potential limit V_0 (β_s close to 90°), we suggest using Eq. (24) with an analytical expression for the function $\bar{F}(\beta_s,f)$ which interpolates the exact semiclassical functions $F_f(\beta_s)$ presented in Fig. 2. The function which performs this interpolation in a satisfactory way was found to have the form

$$\bar{F}(\beta_s,t) = 2\pi \ln\left(\frac{1+\sin\beta_s}{\cos\beta_s + a_1\phi_1 + a_2\phi_2}\right) \quad , \tag{26}$$

where

$$\phi_1 = 1.8 \sin^{36}\beta_s - 0.8 \sin^{18}\beta_s \quad , \tag{27}$$

$$\phi_2 = 1.8 \sin^{72}\beta_s - 0.8 \sin^{36}\beta_s \quad , \tag{28}$$

$$a_1 = 0.85 \frac{75}{f} \quad , \quad a_2 = 0.5 \frac{f}{1200} \quad . \tag{29}$$

The formula (26) coincides almost completely with the Landau-Zener function (15), except for a small region of β_s close to 90° where if follows roughly the "exact" $F_f(\beta_s)$ functions. The differences between the interpolation function (26) and the $F_f(\beta_s)$ functions lie mostly in the limits of 20%.

We shall apply now the results of the spherical box exact semiclassical calculation to the case mentioned above for a Cl atom in a Xe cage. According

to the semiempirical treatment of this system [19], the low-energy absorption peak can be described roughly as a transition in the spherical box with radius $R_0 = 1.2$ Å and transition energies $V_c = 3.95$ eV and $V_0 = 3.54$ eV. Substituting these box parameters together with the 50 K average velocity of $v = 150$ m/s into Eq. (23), one obtains $f = 500$, which gives approximately the absorption maximum at $\beta_s = 75°$ and the half-intensity width $55° < \beta_s < 87°$. These values correspond to $\hbar\omega = 3.58$ eV and a line width of $\Delta(\hbar\omega) = 0.15$ eV, which are close to the experimental values $\hbar\omega = 3.73$ eV and $\Delta(\hbar\omega) = 0.16$ eV [19].

ACKNOWLEDGMENTS

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Figure Captions

Fig. 1. Spherical box model. R_0 is the box radius, β_s the seam angle and B_1B_2 the atom trajectory.

Fig. 2. The function G (14) of the Landau-Zener probability (13) and the functions F_f (24) of the exact semiclassical probability (23). The F_f-functions are denoted by values of the parameter f (22).

Fig. 3. Transition probability per time unit as a function of photon energy for the following parameters of the model: box radius R_0 = 1 Å, the difference of the potential values V_c-V_0 = 0.4 eV, coupling energy u = 0.001 eV, and the atom velocities v = 50, 200 and 800 m/s (f = 1216, 304, 76). The transition probability obtained in the Landau-Zener approximation is denoted by L.Z. The exact semiclassical probabilites are denoted by the velocity values.

Fig. 1

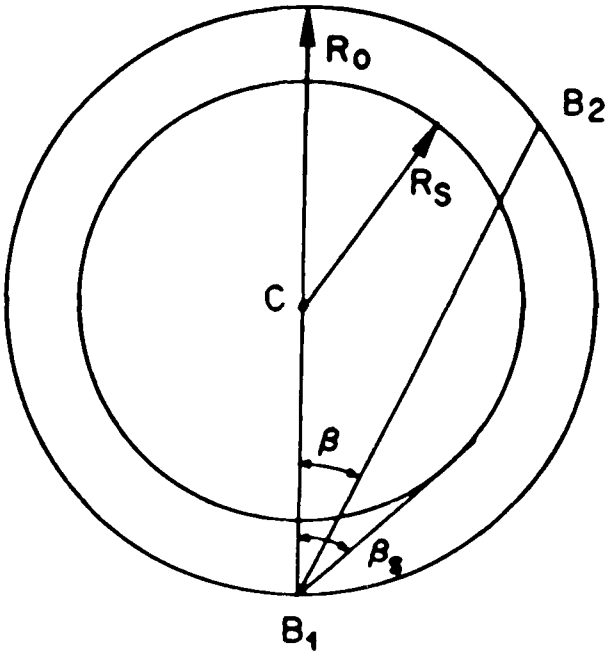

Fig. 2

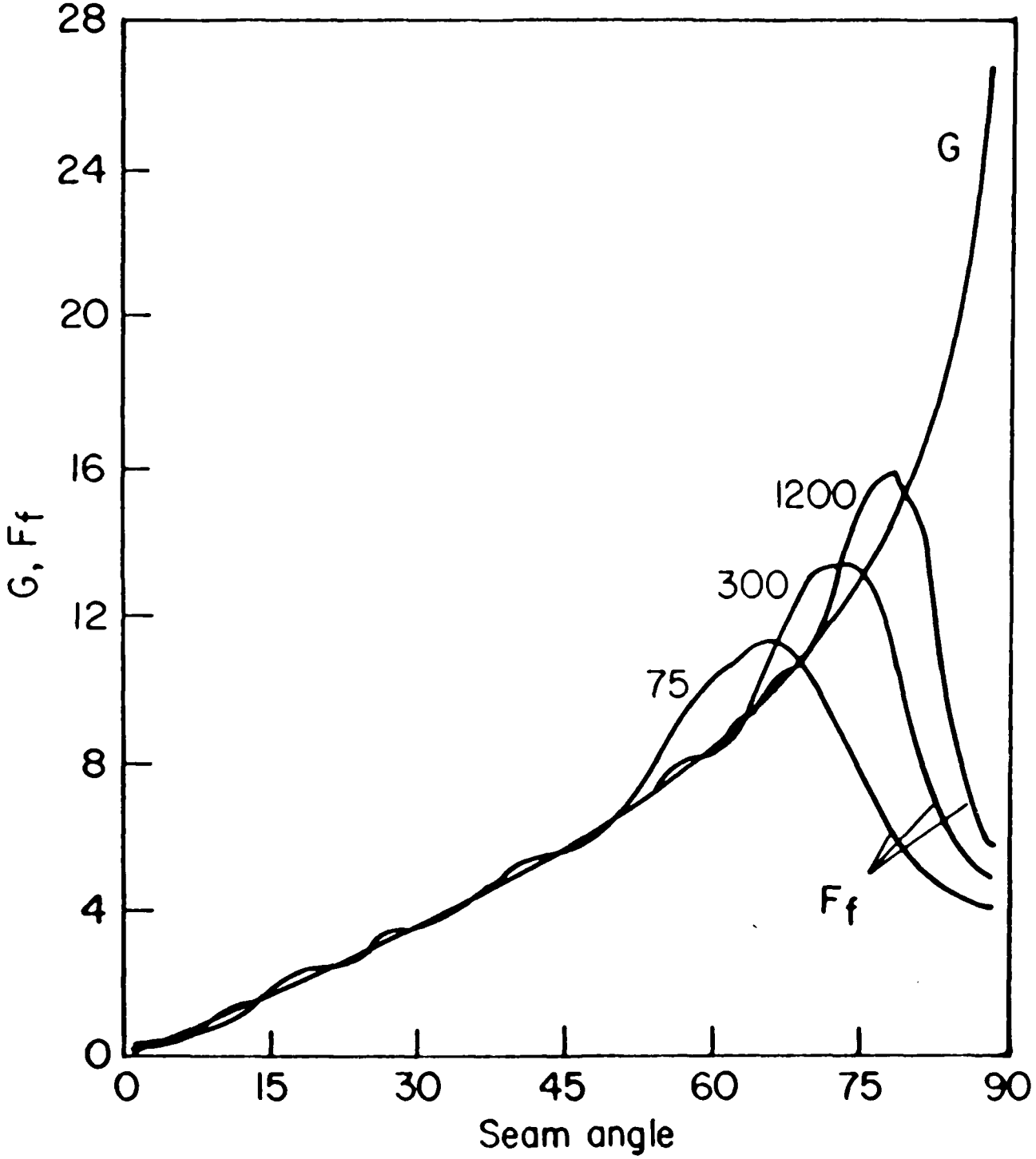
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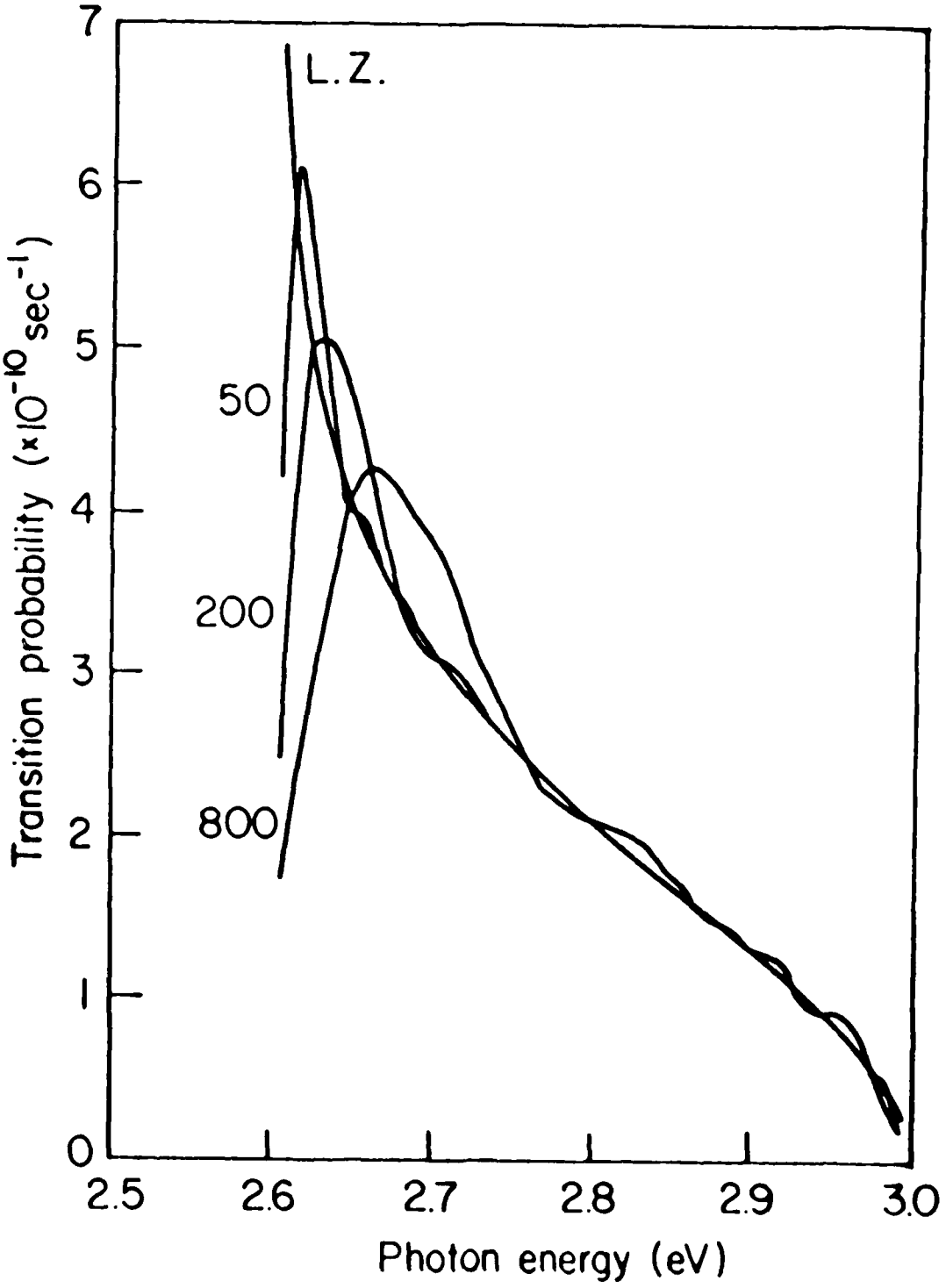
Fig. 3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